

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE*In re* Application of:

Crowther, et al.

Serial No.: 09/955,507

Filed: September 18, 2001

Title: Catalyst Compounds, Catalyst
Systems Thereof and Their Use in A
Polymerization Process§
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BEFORE THE EXAMINER:

Rob Rábago

Group Art Unit No.: 1713

Attorney Docket No.: 1999U033.US-CON

Houston, Texas

July 21, 2003

Commissioner for Patents
P. O. Box 1450
Alexandria, VA 22313-1450**APPEAL BRIEF**

Sir:

This is an Appeal to the Board of Patent Appeals and Interferences from the non-Final Rejection mailed May 2, 2003.

I) Real Party in Interest

Inventors' assignment recorded December 13, 2001, at reel 012359, frame 0004, shows the assignee as Univation Technologies, LLC. This entity is the real party in interest.

II) Related Appeals or Interferences

Applicants are unaware of any related Appeals or Interferences.

III) Status of the Claims

Claims present in the above referenced case are 4-12 and 40. The claims as currently constituted are found in section X. (Appendix) attached to this Brief.

IV) Status of the Amendments

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An amendment under 37 CFR § 1.111, is being submitted concurrently with this Brief to place the claims in better condition for Appeal. The Claims in Section X (Appendix) reflect the claim amendments submitted in this concurrent Paper.

V) Summary of the Invention

This invention relates to a catalyst compound, catalyst systems thereof and to their use in a polymerization process. The catalyst compound is, or a cyclic germanium bridged bulky ligand metallocene-type compound. In one embodiment, this cyclic germanium bridged bulky ligand metallocene-type catalyst compound is activated or combined with an activator to form a catalyst system. In one embodiment, the invention provides for a process for polymerizing ethylene alone or in combination with one or more other olefin(s) in the presence of a catalyst system of a cyclic germanium bridged bulky ligand metallocene-type catalyst compound, which is combined with an activator.

It is highly unusual in the art that a combination of catalyst compound structures provide the benefits to a particular polymer product that each bring separately. Thus, it was surprising and totally unexpected that the cyclic germanium bridged bulky ligand metallocene-type catalyst compounds would produce, in a polymerization process, a polymer having both a high melt index ratio (MIR) and melt strength (MS).

Generally, bulky ligand metallocene-type catalyst compounds include half and full sandwich compounds having one or more bulky ligands bonded to at least one metal atom. Typical bulky ligand metallocene-type compounds are generally described as containing one or more bulky ligand(s) and one or more leaving group(s) bonded to at least one metal atom. In one embodiment, at least one bulky ligand is η -bonded to a metal atom, or η^5 -bonded to the metal atom.

For purposes of discussion, a cyclic germanium bridge is one in which the germanium element(s) serves as the bridging element(s) between at least two bulky ligands and "cyclic" refers to the atoms forming a ring or ring system containing the germanium element(s).

The bulky ligands are generally represented by one or more open, acyclic, or fused ring(s) or ring system(s) or a combination thereof. In these bulky ligands, ring(s) or ring system(s) are typically composed of atoms selected from Groups 13 to 16 atoms of the Periodic Table of Elements, the atoms may be selected from the group consisting of

carbon, nitrogen, oxygen, silicon, sulfur, phosphorous, boron and aluminum or a combination thereof. The ring(s) or ring system(s) may be composed of carbon atoms such as, but not limited to, those cyclopentadienyl ligands or cyclopentadienyl-type ligand structures or other similar functioning ligand structure such as a pentadiene, a cyclooctatetraenediyl or an imide ligand. The metal atom may be selected from Groups 3 through 15 and the lanthanide or actinide series of the Periodic Table of Elements. Or the metal may be a transition metal from Groups 4 through 12, or 4, 5 and 6, or the metal is from Group 4.

In one embodiment, cyclic germanium bridged bulky ligand metallocene-type catalyst compounds are represented by the formula:



where M is as described above or M is zirconium, hafnium or titanium. The bulky ligands, L^A and L^B , are open, acyclic, or fused ring(s) or ring system(s) such as unsubstituted or substituted, cyclopentadienyl ligands or cyclopentadienyl-type ligands, heteroatom substituted and/or heteroatom containing cyclopentadienyl-type ligands. Non-limiting examples of bulky ligands include cyclopentadienyl ligands, indenyl ligands, benzindenyl ligands, fluorenyl ligands, octahydrofluorenyl ligands, cyclooctatetraendiyl ligands, azenyl ligands, azulene ligands, pentalene ligands, phosphoyl ligands, pyrrolyl ligands, pyrazolyl ligands, carbazolyl ligands, borabenzene ligands and the like, including hydrogenated versions thereof, for example tetrahydroindenyl ligands. In one embodiment, L^A and L^B may be any other ligand structure capable of η -bonding to M, or η^3 -bonding to M, or η^5 -bonding to M. In another embodiment, L^A and L^B may comprise one or more heteroatoms, for example, nitrogen, silicon, boron, germanium, sulfur and phosphorous, in combination with carbon atoms to form an open, acyclic, or a fused, ring or ring system, for example, a hetero-cyclopentadienyl ancillary ligand. Other L^A and L^B bulky ligands include but are not limited to bulky amides, phosphides, alkoxides, aryloxides, imides, carbolides, borollides, porphyrins, phthalocyanines, corrins and other polyazomacrocycles. Independently, each L^A and L^B may be the same or different type of bulky ligand that is bonded to M.

VI) Issues

- A. Whether claims 4-12 and 40 are indefinite under 35 USC § 112, second paragraph;
- B. Whether claims 4, 5 and 7-12 are rendered obvious under 35 USC § 103(a) over U.S. 5,491,205 (Langhauser);
- C. Whether claims 4, 5, and 7 are rendered obvious under 35 USC § 103(a) over "Oranometallics" 1994 (Chen) optionally in view of U.S. 6,057,408 (Winter I) or 5,532,396 (Winter II);
- D. Whether claims 4, 5, 7-12 and 40 are rendered obvious under 35 USC § 103(a) over Winter I.

VII) Grouping of the Claims

The claims stand or fall together.

VIII) Argument**Issue A**

Applicants do not agree with the Examiner's restrictive interpretation of "cyclopentadienyl-type" stated in the Office Action dated May 2, 2003. Further, Applicants reject the Examiner's characterization of a lack of definition of "other similar functioning ligand structure(s)".

Applicants have pointed out:

i) that the Specification provides the person of skill in the art ample disclosure to structures other than the restrictive interpretation "cyclopentadienyl-type ... is deemed to be a ligand which includes in its structure an anionic aromatic C₅ core ..." as stated by the Examiner.

To wit: at page 4, line 22, starting with cyclooctatetraenediyl ligands and ending on line 26. Further at page 4, line 36 c/o page 5, line 25, substitutions which can be made on the ligands are clearly delineated;

ii) that the Examiner's own reference to Hawley's Chemical Dictionary contemplates other ligand types.

To wit: Hawley's, as pointed out in the Interview of April 23, 2002, lists among other references "... See ... uranocene." Applicants have provided 2 references to support their position that those of skill in the art will understand the terminology used in the

present Specification. These references include JACS Dec. 18, 1968, where the authors discuss the mass spectra of uranocenes and conclude "This mass spectrum is completely consistent with an assigned sandwich-complex structure of the ferrocene type with a planar eight-membered rings above and below a central uranium atom in a D_{8d} or D_{8h} arrangement." This reference identifies uranocene (See the title "Bis(cyclooctatetraenyl)uranium (Uranocene) ...") and shows that metallocene-type compounds are not restricted to cyclopentadienyl ligands or those having a C_5 core. There is also a mixed ligand example in Kroon, et al., JOMC, 1970, 25, 451-454 (See p. 452, second paragraph, second sentence.) "The Ti atom is 'sandwiched' between the cyclopentadiene and the cyclooctatetraene groups which are both planar within the experimental error."

iii) Other references provide support for additional ligand types. In Britovsek, et. Al., Angew, Chem. Int. Ed., 1999, 38, 428-447 (See p. 440, the paragraph just above section 6.) "Replacement of both Cp groups of a Group 4 metallocene with isolobal, dianionic imido ligands enables high-valent chromium (VI) compounds with metallocene-like characteristics to be accessed." Also, in Gleiter, et. Al., Organometallics, 1989, 8, 298-306 (See the title description "Half-Open Metallocenes" (i.e. metallocenes containing open pentadienyl ligands). Also, the first sentence of the body at the bottom of p. 298. "Recently, the physical and theoretical natures of pentadienyl groups, and their metal complexes, have been of interest, especially relative to their better known cyclopentadienyl counterparts." Also, note the comparative drawings on p. 299 showing the structural relationship between cyclopentadiene and pentadiene ligands and the various combinations that can be observed. There is clear evidence that "metallocene-type" compounds can be composed of "bulky ligands" such as cyclooctatetraenyl, imido, and pentadienyl ligands. Copies of each of these documents are attached as Appendix II.

iv) Additional language was added in a previous response to support the use of the term "bulky ligands" so this issue has already been addressed.

Accordingly, Applicants have provided substantial support for their terminology, and Applicants believe that:

- a) they have sufficiently delineated the contemplated compounds in the Specification on pages 3-11 (paragraphs [011] - [030]), for persons of skill in the art; and

- b) they have shown that as far back as 1968, those of skill in the art understand the meanings of "metallocene-type" stated in the present Specification.

Applicants agreed in the April 23, 2003 Interview to place a statement on the Record that the scope of Applicants' claims, with specific regard to "metallocene-type", would be interpreted by the disclosure of the Specification. Such a statement is included in the response under 37 C.F.R. § 1.111 filed concurrently with this Brief.

The Examiner maintains that the claims are indefinite. The MPEP at § 2173.02 states:

"Definiteness of claim language must be analyzed, not in a vacuum, but in light of:

- (A) The content of the particular application disclosure;
- (B) The teachings of the prior art; and
- (C) The claim interpretation that would be given by one possessing the ordinary level of skill in the pertinent art at the time the invention was made.

Applicants believe that (A), (B) and (C) have been satisfied. When coupled by Applicants' statement that their claims are to be interpreted in light of the complete disclosure and especially in light of paragraphs [011] – [030], Applicants believe they have discharged their duty that the claims particularly point out that which Applicants regard as their invention.

Accordingly, Applicants request reversal of the holding of indefiniteness and withdrawal on the record of the Examiner's restrictive view and holding regarding the terminology.

Issue B

Applicants have amended the claim to include the terminology "wherein the 2 R's are joined ...". Such terminology excludes the spiro compounds of Langhauser.

Applicants respectfully request reversal of the obviousness Rejection over Langhauser.

Issue C

Chen suggests at page 749 second column, above "acknowledgements", "unfortunately it [the catalyst compound] is unstable above 50° C and its performance as a catalyst under manufacturing conditions cannot be assessed."

Lack of stability in a temperature region where polymers are generally made serves as an antimotivational signpost for the skilled person to do any further work with

the catalysts of Chen. Applicants are claiming "A catalyst system for polymerizing ethylene alone or in combination with one or more olefin(s),...", so the skilled person, represented by the inventors would not be motivated to use Chen for arriving at the presently claimed invention.

Furthermore, the motivation to combine must flow from the references themselves, or from extrinsic evidence. Chen's statement about Chen's compound being unstable would not motivate the skilled person to add a support unless adding a support would change or improve the tendency towards instability. There is no evidence of record, showing that supported catalysts of the Chen type have or show improved stability compared to their unsupported counterparts. If such evidence exists, Applicants respectfully request that it be made a part of the record.

In fact, the Examiner states that the motivation to combine a support with Chen's disclosure because "... for obtaining established benefits, including improved activity, facilitation of pre-polymerization, etc. " missing from this analysis of support benefits is the type of evidence discussed above, that being a showing that supported catalysts of the Chen type show improved stability on supports. (*In re Napier*, 55 F.3d 610, 613, 34 U.S.P.Q.2d 1782). The other question that must be raised is absent evidence of improved stability for such catalysts, the skilled person would have to conclude that what one now has with the combination suggested by the Examiner, is an unstable catalyst on a support.

Absent the motivation to combine, or absent evidence as has been requested, no obviousness exists with this combination of references and Applicants request reversal of the Examiner's holding.

Issue D

Applicants respectfully point out that in Winter's examples 26 and 28, as noted by the Examiner, while exemplifying germanium, do not contain a cyclic bridge, nor are they supported. Absent a cyclic bridge or support, the list of "Ms" and "Rs" at Col. 3, lines 56-63, and the list of supports at Col. 11, lines 45-53, pointed to by the Examiner, together with Winter's examples 26 and 28, constitutes at best, an impermissible "obvious to try" Rejection. Further, there must be some motivation in Winter to incorporate all three elements, and no such motivation, save for what Applicants consider "obvious to try", has been elucidated by the Examiner. The basis of an obviousness rejection must be more than a collection of elements disclosed, rather the motivation to combine has to be found

in the evidence of record. No valid motivational theory has been put forth, and Applicants believe that the only motivational factor in the present instance is Applicants' claims themselves, an impermissible rationale for combination.

Absent such motivation Applicants request reversal of the Examiner's holding.

IX) In Conclusion

Given the facts:

- A) that Applicants have adequately described "metallocene-type", such that persons of skill in the art understand the scope of the claims;
- B) that Applicants have amended the claims as suggested by the Examiner to obviate Langhauser;
- C) that there is no motivation to combine Chen, optionally with Winter I or Winter II; and
- D) that Winter I does not provide sufficient guidance to combine disparate parts of Winter I's disclosure to reach Applicants' claimed combination;

Applicants urge reversal of all Rejections.

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CERTIFICATE OF FACSIMILE TRANSMISSION UNDER 37 CFR 1.8(a)

I hereby certify that this paper is being facsimile transmitted to the United States Patent and Trademark Office
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Appendix I

Claim 4. (Currently Amended) A catalyst system for polymerizing ethylene alone or in combination with one or more olefin(s), comprising a cyclic germanium bridged bulky ligand metallocene-type catalyst compound and an activator, wherein the cyclic germanium bridged bulky ligand metallocene-type catalyst compound is represented by the formula:



where M is a Group 3 to 7 transition metal, each of L^A and L^B is an unsubstituted or substituted, cyclopentadienyl ligand or cyclopentadienyl-type bulky ligand bonded to M; $(R'GeR')_x$ is a cyclic bridging group bridging L^A and L^B , and the two R's are joined to form a cyclic ring or ring system with Ge; independently, each Q is a monoanionic ligand, or optionally two Q's together form a divalent anionic chelating ligand; and where n is 0, 1 or 2 depending on the formal oxidation state of M, and x is an integer from 1 to 4 and wherein the catalyst system is supported.

Claim 5. The catalyst system of claim 4 wherein one of L^A or L^B is a substituted cyclopentadienyl or a substituted cyclopentadienyl-type bulky ligand.

Claim 7. The catalyst system of claim 4 wherein x is 1.

Claim 8. The catalyst system of claim 4 wherein the cyclic germanium bridged bulky ligand metallocene-type catalyst compound is represented by the formula:



where M is a Group 4, 5, 6 transition metal, L^A and L^B are bonded to M and are different, L^A and L^B are selected from the group consisting of unsubstituted or substituted, cyclopentadienyl ligands or unsubstituted or substituted, cyclopentadienyl-type bulky ligand; $(R'GeR')_x$ is a cyclic bridging group bridging L^A and L^B , and the two R's are joined to form a cyclic ring or ring system with Ge; independently, each Q is a monoanionic ligand, or optionally two Q's together form a divalent anionic chelating ligand; and where n is 0, 1 or 2 depending on the formal oxidation state of M, and x is an integer from 1 to 4.

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Claim 9. The catalyst system of claim 8 where x is 1.

Claim 10. The catalyst system of claim 8 wherein L^A and L^B are substituted or unsubstituted cyclopentadienyl rings.

Claim 11. The catalyst system of claim 8 wherein a least one of L^A and L^B is a cyclopentadienyl ring.

Claim 12. The catalyst system of claim 8 wherein L^A is a substituted cyclopentadienyl ring.

Claim 13. The catalyst system of claims 4 or 8 where the cyclic germanium bridged bulky ligand metallocene-type catalyst compound is selected from one of the group consisting of cyclotrimethylenegermyl(tetramethyl cyclopentadienyl) (cyclopentadienyl) zirconium dichloride, cyclotetramethylenegermyl (tetramethyl cyclopentadienyl) (cyclopentadienyl) zirconium dichloride, cyclotrimethylenegermyl(tetramethyl cyclopentadienyl) (2-methyl indenyl) zirconium dichloride, cyclotrimethylenegermyl(tetramethyl cyclopentadienyl) (3-methyl cyclopentadienyl) zirconium dichloride, cyclotrimethylenegermyl (tetramethyl cyclopentadienyl) (2,3,5-trimethyl cyclopentadienyl) zirconium dichloride, cyclotrimethylenegermyl bis(tetramethyl cyclopentadienyl) zirconium dichloride, cyclotetramethylenegermyl(tetramethyl cyclopentadienyl) (3-methyl cyclopentadienyl) zirconium dichloride, cyclotetramethylenegermyl bis(tetramethyl cyclopentadienyl) zirconium dichloride, 3,4-dimethylcyclotetra-methyl-3-enegermyl(tetramethyl cyclopentadienyl) (cyclopentadienyl) zirconium dichloride, 3,4-dimethylcyclotetramethyl-3-enegermylbis(tetramethyl cyclopentadienyl) zirconium dichloride, 3,4-dimethylcyclotetramethyl-3-enegermyl(tetramethyl cyclopentadienyl) (2,3,5-trimethyl cyclopentadienyl) zirconium dichloride, 3-methylcyclotetramethyl-3-enegermyl bis(tetramethyl cyclopentadienyl) zirconium dichloride, 3-methylcyclotetramethyl-3-enegermyl (tetramethyl cyclopentadienyl) (cyclopentadienyl) zirconium dichloride, 3-methylcyclotetramethyl-3-enegermyl (tetramethyl cyclopentadienyl) (3-methylcyclopentadienyl) zirconium

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dichloride, o-xylidenegermyl bis(tetra methyl cyclopentadienyl) zirconium dichloride, o-xylidenegermyl(tetramethyl cyclopentadienyl) (cyclopentadienyl) zirconium dichloride, and o-xylidenegermyl(tetramethyl cyclopentadienyl) (3-methylcyclopentadienyl) zirconium dichloride.

Claim 40. The catalyst system of claim 4 wherein the bulky ligands are differently substituted.

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I hereby certify that the correspondence is being facsimile transmitted to: Commissioner for Patents on <u>July 21, 2003</u>		In re Application of <u>Crowther, et al.</u>	
Signature <u>Dorothy Williams</u> Typed or Printed name <u>Dorothy Williams</u>		Application Number <u>09/855,807</u>	Filed <u>September 18, 2001</u>
		For Catalyst Compounds, Catalyst Systems Thereof and Their Use in A Polymerization Process	
		Group Art Unit <u>1713</u>	Examiner <u>Rob Rabago</u>

Applicant(s) hereby appeals to the Board of Patent Appeals and Interferences from the decision of the Examiner.

The fee for this Notice of Appeal is (37 CFR 1.17(b)) \$320.00

☐ Applicant claims small entity status. See 37 CFR 1.27. Therefore, the fee shown above is reduced by half, and the resulting fee is: \$

☐ A check in the amount of the fee is enclosed.

☒ Payment by Credit card. Form PTO-2036 is attached.

☐ The Director has already been authorized to charge fees in this application to a Deposit Account. I have enclosed a duplicate copy of this sheet.

☐ The Director is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. . I have enclosed a duplicate copy of this sheet.

☐ A petition for an extension of time under 37 CFR 1.136(a) (PTO/85/22) is enclosed.

I am the
☐ applicant/inventor.
☐ assignee of record of the entire interest.
 See 37 CFR 3.71, Statement under 37 CFR 3.73(b) is enclosed.
 (Form PTO/85/90)
☐ attorney or agent of record
 Registration number
☒ attorney or agent acting under 37 CFR 1.34(a).
 Registration number if acting under 37 CFR 1.34(a).
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